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for the Measurement of Radioactivity in
the Ocean

Primeneniye Glubokovodnogo Gamma-Radiometra
dlya Izmereniya Radioaktivnosti v Okeane

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The Use of a Deep-Water Gamma Radiometer for the
Measurement of Radioactivity in the Ocean

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The total natural radioactivity (r/a) of the ocean is approximately equal to $5 \cdot 10^8$ curie and is caused in the main by the following isotopes: K^{40} , Rb^{87} , U^{238} , U^{235} , Th^{232} , Ra^{226} [1].

In addition, there can be found in sea water natural radioactive isotopes of cosmic origin such as C^{14} , T , Be^{10} , Be^7 , Na^{22} , P^{32} , P^{30} , S^{35} , but their part of the total activity of the ocean is small.

The major isotope causing significant r/a of the ocean [1] is the isotope of potassium-40 ($T_{1/2} = 1.31 \times 10^9$ years, β (90%) = 1.32 mev., γ (10%) = 1.459 mev.).

Lately, in connection with the carrying out of tests of atomic weapons and with the increase in the practical use of atomic energy, in the waters of the World ocean there has appeared a significant amount of r/a isotopes.

We cannot here go into any degree of detail on the matter of contamination of the oceans by r/a isotopes; we will note only that

in the last few years questions connected with this problem were raised several times (first and second conferences for the peaceful use of atomic energy in Geneva, 1956-1958), the section of the Pacific Ocean oceanographic congress in Bangkok in 1959, the Conference on isotope burial in Monaco in 1959 and others. One of the conclusions in the proposition of the non-local character of the contamination of the ocean, according to which r/a isotopes can as a result of oceanic currents or of air mass translocations be carried great distances from the site of the atomic blast [2].

The distribution of r/a in sea ~~water~~ is usually studied by radiochemical methods with the concentration and chemical separation of its isotopes [3] ~~prior to measuring~~. Together [page 171] with this exact, sensitive, but laborious method, attempts were undertaken to measure directly the increased content of r/a in sea water.

A whole line of instruments for the determination of r/a in surface layers was created at the Oceanographic Institute in the U.S.A. [1]. The pickup units of these instruments, the major part of which were gas counters, were connected by a cable to the recording system on board the ship. These instruments allowed the determination of the gamma background on the surface of the water and at a small depth during movement of the ship.

B.A. Nelepe [4] employed a radiometer using scintillation data units for measuring r/a of Antarctic and North Atlantic waters. The measurements were done by him in depths up to 150 meters while

the counter pulses were able roughly to discriminate energies by means of the counting apparatus (FS-10000 "Flocks") aboard the research ship "Ob". V.N. Lavrenchik and G.N. Sofiyev [5] carried out several measurements of the r/a in the waters of the Indian Ocean in depths up to 1000 meters, there being received on a 5C channel analyzer several emission spectra.

In all instances the length of the transmission cable limits the possibility of measurements at great depths.

At the same time, the direct and fast measurement of r/a at depths greater than 1000 meters present significant interest not only for the elucidation of the character of the distribution of r/a in the ocean but also for the elucidation a whole series of hydrological questions (the limits and directions of currents, the origin of oceanic masses, etc.).

Of special importance is the elucidation of the question of the behavior of r/a in deep-water depressions of the ocean since at the present time in connection with the question of burial of r/a wastes of nuclear transformations, there has been brought forward the proposition of the possibility of carrying out the above burial in the maximal depths of the ocean. The final answer to that question can be given by the direct study of the distribution of r/a isotopes at great depths.

Apparatus for the deep-water measurement of r/a did not exist. In the present work the deep-water radiometer RA7-1 is described and several results of the measurement of r/a in the Indian Ocean

are reported. The work was done in 1959-1960 in Moscow and on board the research ship "Vityaz'" during its 31st run.

Description of the Apparatus

The small number of underwater radiometers described in the literature are characterized by the fact that only the radiation detector is lowered by a cable (or cable-hawser) to the given level of measurement. The whole receiving apparatus is located aboard the ship. [Page 172]

The value of such a method is:

- a) the possibility of taking emission spectra (in the case of scintillation detectors).
- b) the continuous measurement and the control of the apparatus;
- c) the convenience and simplicity of measurements at shallow depths.

However, systems of such a type also have serious deficiencies: a severe complication of work with use of a cable-hawser of great length, the lack of dependability of its operation at great pressures and also a sharp increase in the work of measurement since the use of a cable-hawser calls forth the necessity of the installation of a special deep-water winch (of the GOL or trawling types), the work of which is connected with great expenditures of time. Since the use of a cable-hawser of great length will be justified only upon the transmission of emission spectra (and not upon the simple count of r/a), its use will lead to further complication of the receiving apparatus (multi-channel analyzer).

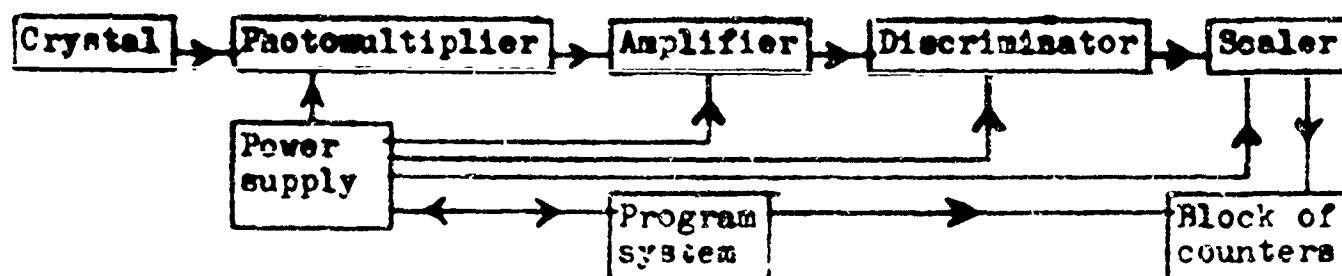


Figure 1. Block diagram of deep-water radiometer.

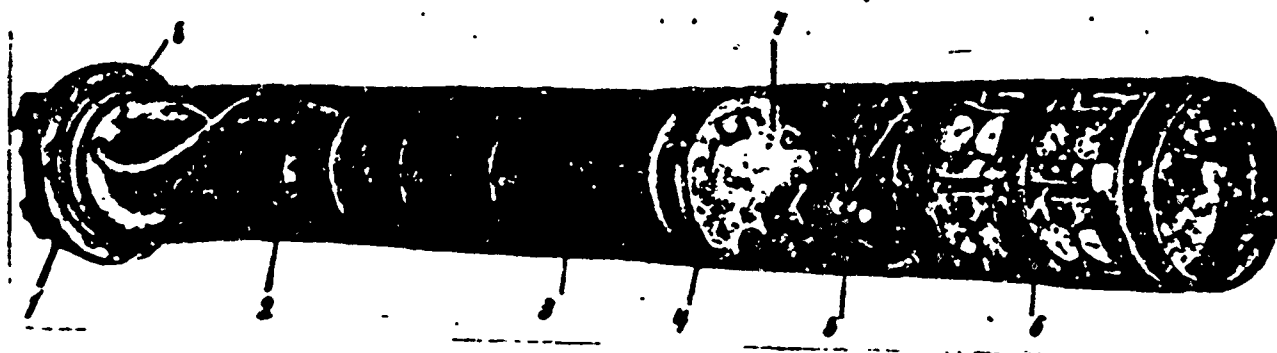


Figure 2. General appearance of radiometer (without outer shell).
 1-flange; 2-sleeve of photomultiplier and crystal; 3-battery;
 4-amplifier, discriminator, scaler device; 5-program system; 6-block
 of counters; 7-automatic power switch; 8-gasket.

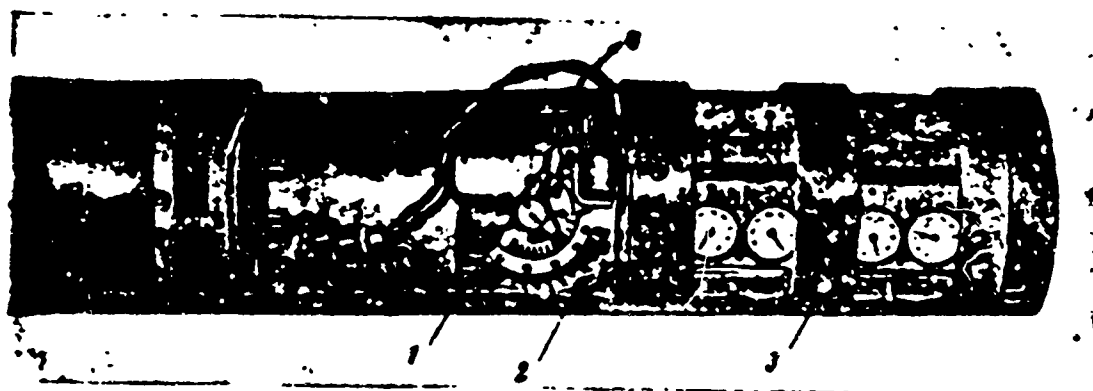


Figure 3. Block of counters and program system.
 1-relay; 2-step-by-step switch; 3-block of counters; 4-electric
 clock.

Another constructive answer is possible: the placing of the whole receiving apparatus together with the data set in an autonomous depth shell.

With this: a) the lowering of the radiometer can be done with the normal hydrological winch (of the "Okean" type) on a hawser, which does not place any limitation on the depth of measurement; in view of the absence of a lead cable, the guarantee of hermeticity of the apparatus is increased; the noiseproof quality is increased and induction on the transmitting cable is removed; the simplicity of operation is increased. Disadvantages of apparatus of such a type are the impossibility of control of operation of the apparatus when submerged and the delay in the receipt of information until the radiometer surfaces.

However, the advantages of such an apparatus for the measurement of r/a at great depths were so obvious that they determined the construction of the RAG-1 radiometer described. [page 174]

A block diagram of the parts of the deep-water radiometer is presented in figure 1, the general appearance of the apparatus and its separate blocks and a radiotechnical schematic are presented in figures 2-4.

An NaI crystal, 30x10 mm. in size was used in the deep-water radiometer.

The photomultiplier of the FEU-29 type is powered by a battery. The voltage on the photomultiplier is about 1000 v. while the

sensitivity (according to the specifications) is equal to 100 amp/lumen. From the anode of the photomultiplier, impulses impinge on a twin emitter repeater which is assembled from P-402 diffused transistors. The time leading constant of the photomultiplier is 6 microsec.

The basic requirements for the amplifier of the deep-water radiometer are economy and stability of function. This predetermined the choice of design to transistors with feedback coupling and thermocompensation. The amplifier is constructed from transistors with various types of conductance. For the strengthening of signals of negative polarity P-15 (p-n-p) germanium triodes are used, and for positive, P-103 (n-p-n) silicon triodes are used. An amplifier of this design has the greatest stability. The first two cascades before the discriminator on the D-2E diode have a coefficient of amplification of about 100, which at the cutout voltage of the discriminator of 1.6v. insures the counting of pulses above 16 mv. In both cascades, feedback coupling and thermostabilization are employed by the insertion of the emitter and discriminator resistances into the base resistances. After the discriminator follows the 2-cascade amplifier with a coefficient of amplification on the order of 800. This amplifier is not thermostabilized. The load on the second cascade is the step-up transformer with a transformation coefficient of 1:6. The output pulse of the amplifier has a positive polarity and a duration of 100 microsec. This pulse is fed to the normalizer, which is assembled from a channelless thyatron [hot-cathode grid-controlled gas tube]. From the normalizer, pulses of 200 microsec. duration and 40v. amplitude are fed to the scaler device.

The introduction of a scaler device into the design of the deep-water radiometer allows the measurement of oceanic r/a of a wide band of activity. A choice of counting $x1$, $x2$, $x8$, $x16$, and $x32$ is allowed which gives a possibility for making errors from uncounted pulses considerably less than the statistical error. Moreover, the scaler device allows calibration of the radiometer with a standard of comparatively high activity. The scaler device is assembled from 5 identical MTKh-90 [MTX-90] $\times 2$ counting stages. The limiting time for the counting of periodic pulses is equal to 2 msec. The scaler device works on the output stage of a mechanical counter, assembled also from MTKh-90.

The recording part of the radiometer (figure 4) is a block of 10 mechanical SB-100-M counters connected to the scaler device in a specific sequence by the program system. [page 175]

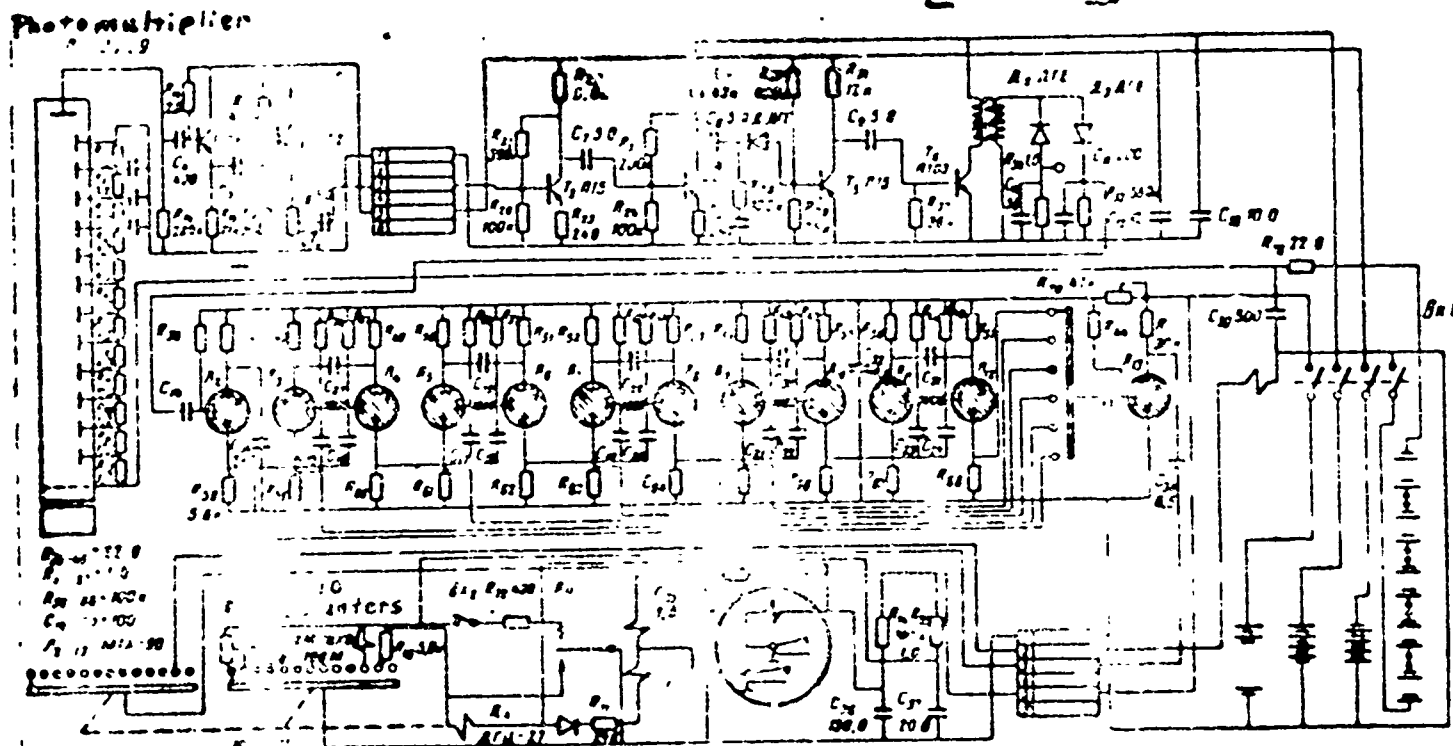


Figure 4. Radiotechnical schematic of the apparatus. page 176

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The program system includes an electric clock, a polarized relay, and a SHI-11 [W4-11] step-by-step switch. The clock contacts close the power circuit of the windings of the polarized relay, the operation of which causes a turn of the step-by-step switch to the next position and, consequently, a switching of the counter. The period of operation of the program system is 10 min. ± 0.2 sec. Work is possible in one of two regimes: switching of the counters every 10 min. and switching with 10 min. pauses between measurements. Between the switching on of the power and the switching on of the first counter is a period of 10 min. Upon switching of the step-by-step switch into the last (11th) position, the electromagnetic shut-out operates and the radiometer circuit is turned off completely.

6

The power block consists of four dry batteries. A high voltage battery is built from a disk coil of GE-100 batteries and divided into five sections to reduce self-discharge. The capacity of the battery is 0.05 amp/hr which with a resistance load of 100 mohm gives a service period of 5000 hours. The power battery of the scaler device and automatic equipment is composed of BAS-G-60-L-2.3 disk coil batteries and has a capacity of 2.3 amp/hr. at a voltage of 150v. The period of service of this battery is 1600 hours. The power batteries of the amplifier (+6v. and -6v.) are made of disk coils of BAS-G-2.1 batteries and have a capacity of 2.1 amp/hr. Current is used from both batteries at 1 mamp, which gives a period of service of 2100 hours. Thus the period of work of the deep-water radiometer on one set of batteries is in fact limited only by the

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storage period of the batteries. This is of great significance since during the time of one measurement (200 min.), the power voltages cannot change significantly at all. A radiotechnical schematic of the radiometer is given in figure 5 [sic].

In the course of work on RAG-1 it was established that the slightly modified radiometers "Svet-3" and "Kristall" during extensive continuous operation do not supply stable readings without readjustment of the anodal and filament voltages, which excludes their use as a detecting unit in automatic radiometers.

For the study of the possibility of direct measurement of the beta activity of sea water several measurements were done with a p-terphenyl crystal placed on the outside of the radiometer. The light from the crystal passed through a special light conductor mounted in the flange. The results of the experiment showed that upon increase in pressure with depth, optical contact between the crystal and the light conductor is broken and the count is stopped.

The body of the radiometer is made of steel tubing with $\phi_{\text{outer}}=200\text{mm.}$, $\phi_{\text{inner}}=140\text{mm.}$ and length=1400mm. The weight of the radiometer in its body is about 200 kg. The outer flange with the ring for the attachment [page 177] of the hawser is welded to the body. The lower flange is removable. Onto it is attached a brass sleeve to which all blocks of the radiometer are secured. The connections between the blocks are made with six-contact connectors. The flange is secured to the body with stainless steel pins. Hermeticity of the joint is effected by a self-packing gasket rated for pressures up to 1000 atm.

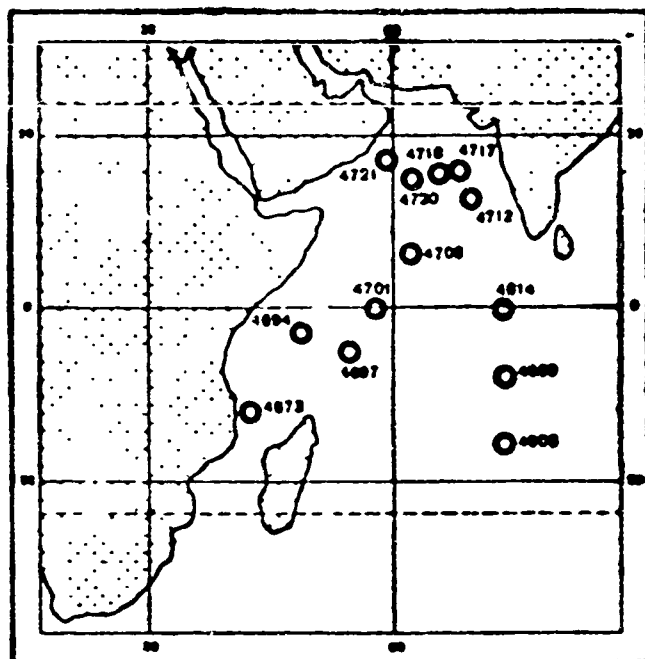


Figure 5. Chart of the 31st run of the research ship "Vityaz"

The circles with numbers indicate the oceanographic stations where the radiometric surveys were made (see [6] for station coordinates).

Calibration of the radiometer was done in a sufficiently large circular section tank ($\phi=5.15$ m, $N=4\text{m}$), volume of 80.2 m^3 and having a cement covering above. For the exclusion of the effect of the cement's own radiation, the radiometer was placed at a distance of 1.5 m from the walls and bottom of the tank.

The isotope K^{42} was chosen for the calibration process. It was chosen as an isotope with a short half-life (12.44 hours), with a type of radiation resembling that of natural K^{40} (K^{42} -1.51 mev., K^{40} -1.46 mev.), and also with simplicity of production (by p, γ reaction).

The radiochemical purity of the preparation was determined by its half-life (during calibration) and by its gamma-spectrum taken

on a 100-channel analyzer. [page 178] The salt containing K^{42} was dissolved in 25 ml. of water and was poured through a funnel and glass tube into the bottom of the water filled tank.

The equal distribution of the added radioactivity throughout the whole volume of the tank was effected by the energetic mixing of the water by a stream of compressed gas (nitrogen).

Measurements were taken over the course of 6 days: the period of complete disintegration of the isotope.

With the exception of the first few hours of measurement, when an excessively high level of activity caused errors in the apparatus intended for slow speed - pulse arrival, the data for the pulse values were close in magnitude. Thus it was established that one pulse recorded by RAG-1 corresponds to 31 gamma quanta/liter or to 0.348×10^{-10} curie/liter for K^{42} or 0.725×10^{-10} curie/liter for K^{40} .

Procedure

The procedure was as follows:

1. Development of a schedule measurements. The radionometer allows two types of switching of the mechanical counters: consecutively (regime I) or with 10 minute intervals (regime II). The choice of switching regime is determined by the intended goal, i.e. when getting results with maximum statistical accuracy at one level, the most rational is the consecutive switching of counters (regime I). If, however, the measurements are done at various levels, a ten minute interval between switchings is necessary for placement of the radionometer at the assigned level (regime II).

Work regimes I and II differ substantially with regard to the time spent on the whole measurement (100 min. for regime I and 190 min. for regime II). Therefore, in connection with the necessity of saving time, measurements were sometimes taken on various levels using regime I--two on each level. With this, of course, not all 10 counters were used since part of them operated during the raising and lowering of the radiometer.

2. The translocation of the radiometer from one level to another was done strictly by the clock--simultaneously with the release of the time relay of the radiometer, a timer on the deck was turned on. The speed of raising and lowering the apparatus was determined exactly. Before each translocation, the radiometer was held at the level just measured for 1-2 min.

Usual translocations of the radiometer were 1000-2000 m., since the apparatus traveled through these distances in the course of 8-9 min. which prevented the loss of data of one of the counters.

3. Calibration of the apparatus was done before each series of measurements, before each lowering. The delivery of an identical voltage [page 179] to the photomultiplier was controlled by the count of a standard (cobalt-60: 0.56 microcurie) with a 3% accuracy. At the same time the scaler device on the apparatus was set at 1:16. The calibration was done in the laboratory for one hour directly before and after lowering the apparatus. Such a calibration allowed comparison with each other of measurements taken at various levels.

4. ~~Hermetic sealing~~ of the apparatus was done on the deck. The sleeve with the apparatus, having been brought from the laboratory, was fitted into the radiometer body which was closed by the flange and gasket.

Taking into account the high humidity of the air (up to 98% at 32°C) in tropical regions of the run and the possibility of condensation of moisture on the inner parts of the apparatus upon its penetration into a cold layer of water, a desiccant cartridge of silica gel was placed inside the radiometer prior to hermetic sealing. The high humidity also required the holding of the apparatus on deck after finishing the measurements since the opening of the cold apparatus elicited a dangerous moistening of its parts.

5. Data from the mechanical counters was recorded in the laboratory.

6. Work with the RAG-1 radiometer was done with the hydrological winch "Okean" on hawsers of 5.6 and 4.5 mm. Two men worked on the radiometer, one of which was the winch operator. Computation of the true level of measurement was done using the angle of inclination of the hawser.

It was possible to combine the work with the radiometer at deep levels with simultaneous work with other deep-water apparatus (dredger, trawler) from one side of the ship. Measurements at depths up to 220 m. were carried out with simultaneous work of apparatus on the opposite side which allowed a significant saving of time.

Results of measurements of r/a in the Indian Ocean*

Station	Level, m.	Intensity of count, impulses/min.	Station	Level, m.	Intensity of count, impulses/min.
4603**	1000	37.8	4708	100	58.9
	1000	38.7		100	58.2
	2000	36.7		100	65.9
	4000	33.1		100	54.2
	4000	35.7		100	58.0
	4000	32.0		0	105.1
4609**	4000	40.5	4712	200	54.9
	4000	45.0		1000	59.8
	3000	46.2		2500	62.0
	3000	45.5		2500	61.2
	2000	41.4		4020	58.8
	2000	42.7		4020	58.4
4609**	200	37.6	4717	20	64
	200	39.1		214	58
	100	41.5		214	61
	0	72.7		100	62
	0	77.2		100	62
	0	79.1		20	65
	0	74.0	4718	100	58.8
4614**	10	49.1		1000	58.0
	50	45.4		2000	58.0
	100	44.4		3000	56.4
	200	42.2		3500	56.4
	200	39.2		3000	58.0
	100	39.2		2000	59.8
	50	44.5		1000	60.0
	10	45.1		100	60.8
	0	70.2		0	116.8
4694	50	59	4720	100	60.4
	200	62		100	58.4
	200	64		200	60.0
	50	61		200	62.4
4697	200	61.4		100	61.2
	1000	58.4		100	64.4
	1000	58.2		200	61.2
	2500	63.2	4721	100	54.4
4697	2500	64.9		1000	60.8
	4000	58.0		2100	52.4
	4000	62.1		3150	55.6
4701	100	60.1		4270 дно	215.5
	100	57.3	4721	100	57.6
	100	59.7		1000	56.0
	100	60.8		2100	56.0
	100	63.7		3150	55.2
	100	60.7		4000	56.8
	100	59.0		3300	58.0
In laboratory	4708	В лаборатории		1000	64.4
	0	94.8		0	118.2

*Measurement time at each level was 10 min.

**Measurements at stations 4603, 4609, and 4614 were carried out with a different count effectiveness from those at the other stations.

The maximum turbulence during some measurements was 5 on the sea breeze scale.

Measurements of r/a of central and northern parts of the Indian Ocean were done with the RAG-1 radiometer. The oceanographic stations where the work was done are presented on a chart (see figure 5).

The results of the measurements are presented in the table.

Before assessing the data obtained, it should be restated that the value of a pulse of the radiometer described (counting coefficient) is 3.24% (1 pulse corresponds to 31 gamma-quanta/min./l.).

The potassium content of sea water is fairly constant and depends on the total salinity, which changes insignificantly. Thus, for instance, at one of the oceanographic stations [page 180-Table-page 181] (№ 4712), where measurements were being taken, the salinity values changed with depth in the following fashion:

Depth, ...m.....	1...0	1	104	1000	2000	3665
Salinity, ‰.....	35.46	. 36.5	. 35.3	34.87	34.75	

i.e. the difference amounted to about 4%. For sea water, the specific activity of K^{40} was determined as 720 decays/min./l., or 86.4 gamma-quanta/min./l., these variations amount to about 3 gamma-quanta/min./l., which will not be recorded by the radiometer at the given pulse value. Therefore, one can consider the natural K^{40} background of sea water constant.

Then, from the pulse value and from the average reading of all measurements--60.2 pulses/min., we get the average background of the radiometer equal to 57.2 pulses/min.

The scatter obtained from the measurements can be attributed to:

- 1) statistical scatter of measurements of both the sea water and the background of the apparatus itself;
- 2) instability of operation of the apparatus.

Let us evaluate first the instability. It was checked several times. The mean scatter of results after 100 min. of measurement of a standard of 0.56 microcurie did not exceed 1%.

In order to check the dependence of operation of the apparatus on temperature changes, the radiometer was subjected to a multi-hour cooling in the food refrigerator chamber of the research ship "Vityaz'". The temperature was thus changed by 40°C (from +34° to -6°). Continuous measurements were made with the abovementioned standard. Deflections of readings during the temperature lowering were +6%.

It should be indicated also that the temperature of deep layers of the ocean, starting at 1000 m., is fairly constant so that the larger part of the measurements are taken at the same temperature.

Consequently, the working instability of the radiometer apparatus does not exceed 6% (in the direction of decrease of readings), which gives a reading change of 3.6 pulses/min.

Statistical errors of measurement. Taking the mean background of the radiometer as 57.2 pulses/min. and the average level of activity of sea water (for the given radiometer) as 3 pulses/min., we obtain:

$$\frac{\Delta I}{I} = \sqrt{\frac{1 + \frac{2I_{\text{back}}}{I}}{I\tau}} = \sqrt{\frac{1 + \frac{114}{3}}{3 \cdot 10}} = 1.17$$

Statistical error ± 3.5 pulses/min. At the same time the calculated data scatter of the apparatus attributed to statistical measurements and apparatus error is ± 10 pulses/min. [page 182]

The general scatter of data on all readings corresponds with this quantity.

It should be noted that with an activity change of the water by two, i.e. with an activity of 160 gamma-quanta/min./l., the data scatter would be not less than 17 pulses/min.

Thus, the minimal threshold sensitivity of the radiometer is 240 gamma-quanta/min./l.

On not one of the oceanographic stations visited in the measurement survey was the scatter found to be more than 10 p./min. at various levels, therefore it can be assumed that the activity levels do not exceed 200 quanta/min./l.

The dependability of this conclusion and the dependability of the calculated value of the radiometer accuracy are substantiated by the experimental data of the calibration of the apparatus. The specific activity of K^{42} of 1.1×10^{-9} curie/l. was clearly recorded by the radiometer (11 pulses/min. on a background of 66 pulses/min. after 10 min. of measurement) with an accuracy of 10%.

Conclusions

1. There was created an original automatic deep-water gamma-radiometer with a sensitivity threshold of 2×10^{-10} curie, intended for the measurement of r/a at the maximum depths of the oceans of the world.

2. During measurements of the gamma-background of the depths of the central and northern parts of the Indian Ocean in 1960, there was not found any increase in the r/a level exceeding the natural level by more than 2-3 times.

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